

7th International Building Physics Conference

# IBPC2018

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## Proceedings

**SYRACUSE, NY, USA**

September 23 - 26, 2018

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Healthy, Intelligent and Resilient  
Buildings and Urban Environments

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## Understanding swelling of wood through multiscale modeling

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### ABSTRACT

Wood is a well-used building material where the capacity of wood to absorb water leads to swelling and reduced mechanical properties, and also to questions of durability. The origin of the hygroscopic behavior of wood lies at the nanoscale material that composes its cell walls. Using atomistic modeling, we study the hygromechanical behavior of the different polymeric components of wood, separately and in aggregate configurations. We report the coupled effects of water sorption on the hygric and mechanical properties of the S2 cell wall layer and we upscale the findings using a poromechanical framework.

### KEYWORDS

Wood, hygroscopic behavior, swelling, multiscale modeling, atomistic modeling.

### INTRODUCTION

Natural materials, from plant origin, are ubiquitous in our daily environment, where they are exposed to environmental loading like temperature and relative humidity variations. In particular, cellulose-based materials such as wood have been used traditionally and ubiquitously, and are particularly attractive nowadays as they offer sustainable solutions and as they can be a source of inspiration for biomimetic design of new materials. Wood, an orthotropic cellular biomaterial, has the capacity of adsorbing water molecules from the surrounding environment into its hierarchical material structure. This material displays remarkable properties combining low weight, high strength and high toughness, originating from the hierarchical organization of crystalline and amorphous polymers.

Wood is a well-used building material, for structure, façade, windows, indoor finishing and furnishing, etc., where the capacity of wood to absorb water is not only an issue of dimensional stability and modified material properties, but also of durability. In effect, as water molecules are adsorbed into the hydrophilic matrix in the cell walls, the induced fluid-solid interaction forces result in a swelling of the cell walls. The moisture-induced phenomena at cell wall scale are reflected in the hygromechanical behavior of wood at macroscale. Adsorption of moisture in wood, in the hygroscopic range i.e. until around 30% moisture content mass per mass, results in swelling up to 10% volumetrically and reduces considerable its mechanical properties.

We propose a multiscale approach to investigate and understand the hygromechanical behavior of wood. At cell wall material scale, using atomistic modeling, we study the different polymeric components of wood, separately and in aggregate configurations, in terms of the coupled effects of water sorption on hygric and mechanical properties. Then, we upscale the findings using a poromechanical approach. Such properties can then be employed in finite-element modeling to investigate the effects of cellular geometry and ring growth

pattern of wood. The work aims at elucidating the origin of the hygro-mechanical behavior of wood and such complex polymeric materials. In effect, wood is retained as a model hygroscopic material for further understanding of such hygromechanical behavior.

## METHODS

### *Molecular Dynamics (MD) simulations*

#### Materials

Wood cell wall is made of a network of stiff cellulose microfibrils which are embedded within complex matrixing polymers) and the S2 chemical content has been in large part identified. Figure 1 displays the cell wall layers and their components. We aim to reproduce all the main polymers present in the wood S2 layer, namely crystalline cellulose, galactoglucomannan (named GGM below), glucuronoarabinoxylan (referred as xylan below) and the different configurations of guaiacyl lignin.

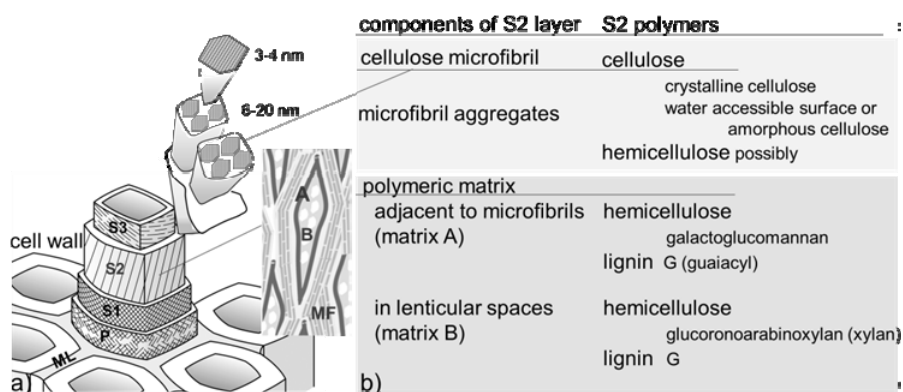


Figure 1. a) Schematic representation of the structure of wood cell wall, with P primary layer, S secondary layer, ML middle lamella (based on Derome et al. 2012), MF microfibril, A matrix close to fibrils, B lenticular matrix; b) components of S2 cell wall layer for a softwood like spruce.

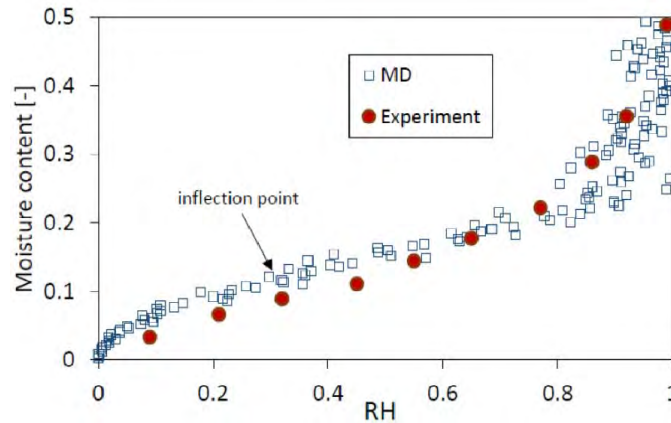
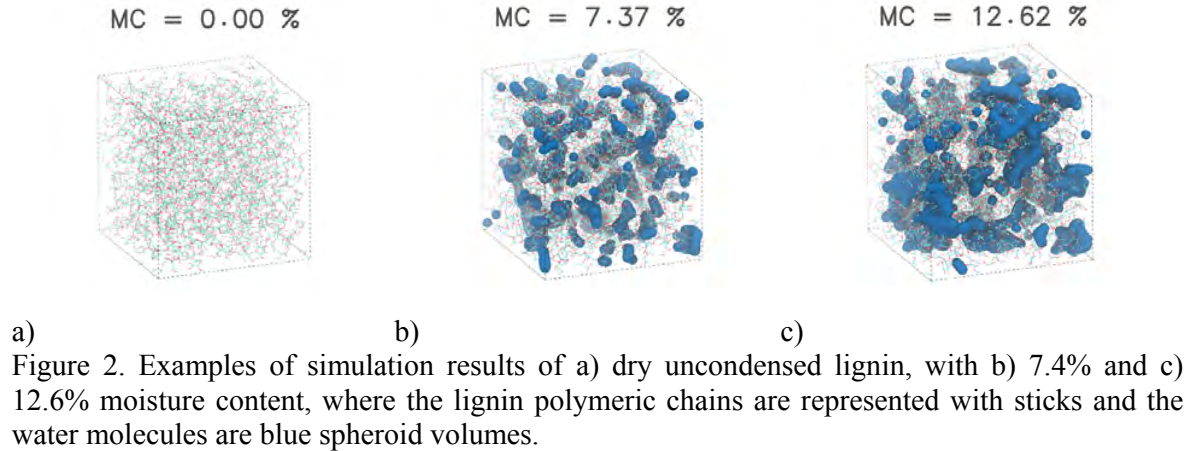
#### Simulation

Molecular Dynamics simulations are carried out using Gromacs software (Hess et al 2008) with leap-frog algorithm for integration of Newton's equations of motion and the Gromos 53a6 united-atom force field (Oostenbrink et al 2004, Lins and Hünenberger 2005). The initial configuration of the polymer is constructed based on the position of atoms in the monomers and the proportion of different monomers in the chains as documented by previous studies. Using the main chemical components of wood, here softwood, the polymeric systems are reproduced, their hygromechanical behavior determined and analyzed. Nanoscale structures or composite systems are built and analyzed. MD results are validated with available experimental data. We note the capacity with this method to control exactly, as input, the polymeric constituents, their polymeric configuration and the configuration of layered or composite systems. Molecular dynamic provides a wealth of data, on which statistical physics is applied and then compared with theoretical and continuum approaches, like thermodynamics, providing a unique window on the fundamental hygromechanical behavior.

#### Tests

In order to study the different responses of the polymers to water, water molecules are introduced one by one to the system and the system let to equilibrate, as shown in Figure 2. We obtain water adsorption curves using One-Step Perturbation method for the determination of chemical potential. Calculated sorption curves, i.e. moisture content versus chemical

potential, are compared with experimental data for validation, as shown in Figure 3. The swelling of the system at each water insertion leads to the determination of the dependence of swelling coefficient on moisture content. Under mechanical loading (compressive or tensile, as desired), i.e. an anisotropic barostat is applied and the pressure specified, the system finds its equilibrium and a corresponding strain can be measured. We can determine the bulk and shear moduli dependence on moisture content. Orthotropic systems are probed by loading in the different directions.



### Upscaling

We use a poromechanical framework as described in Carmeliet et al. (2013) and Kulasinski et al. (2015c).

## RESULTS

We present MD results for the single components, namely long-chained polymers amorphous cellulose AC, GGM, xylan and an uncondensed lignin uLGN. Then we present one mix of amorphous polymers (Matrix), one cellulose crystal embedded in GGM ( $\mu F$ ) and four cellulose crystals embedded in GGM and surrounded by uLGN (S2).

## Swelling

The introduction of water molecules results in a displacement of the polymeric systems leading to material swelling. For the single components and the matrix system, made of amorphous components, swelling is found to be isotropic. Figure 4 shows that volumetric swelling strains vary linearly with moisture content, after an initial nonlinear behavior, the slope of this relationship yielding the swelling coefficient. Swelling coefficients are ranging from 1.07 for S2 to 2.0 for GGM. The different swelling coefficients stem from the combined effects of initial density and porosity, number of hydroxyl sites and bulk modulus, resulting from the different monomers content and arrangement of each system. As noted, the slope at low moisture content is always smaller and nonlinear.

The volumetric swelling of S2 layer is found to be smaller than those of hemicellulose and lignin alone. The crystalline cellulose acts as a stiff skeleton preventing the system from deformation. This effect is particularly pronounced in the longitudinal direction where, due to cellulose covalent bonds, the swelling coefficient is two orders of magnitude smaller ( $\epsilon_z = 2 \times 10^{-3}$  at maximum moisture content) than in the other directions. This indicates that, for the microfibril systems, anisotropy directly results from the presence of crystalline cellulose that restrains most of the deformation in the direction along the crystal.

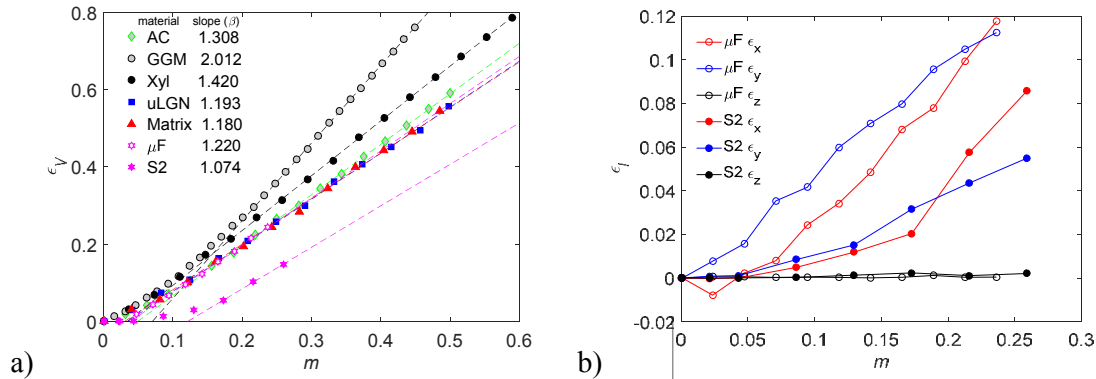


Figure 4. a) Volumetric swelling strain ( $\epsilon_v$ ) of four polymers and one amorphous composite system and of  $\mu F$  and S2, over the full range of moisture content ( $m$ ). b) Orthotropic swelling strains ( $\epsilon_i$ ) of  $\mu F$  and S2, in function of moisture content ( $m$ ) (from Derome et al. 2018).

## Mechanical properties

As moisture content increases, all investigated polymeric systems show a decrease in elastic moduli. In single systems, the decrease of the bulk moduli, determined by imposing tensile or compressive stress, is seen to be in the range of factor 3 or 4. Remarkably the decrease in shear moduli is much more substantial, over one magnitude (Figure 5a, b). These lower shear moduli at higher moisture content are in part due to the presence of water layers offering almost no resistance in shear.

For anisotropic composite systems, Young's moduli are determined in the three principal directions. Results in the two directions  $x$  and  $y$ , perpendicular to the crystal orientation, are very similar. The weaker behavior with increasing moisture content is similar in all directions, but the magnitudes are very different for the behavior parallel to the crystal orientation (Figure 5c, d). The difference between microfibril and S2 is in large part due to the lignin which makes about one quarter of the S2 layer and which was here implemented with a low level of cross-polymerization, and as such is observed to weaken significantly.

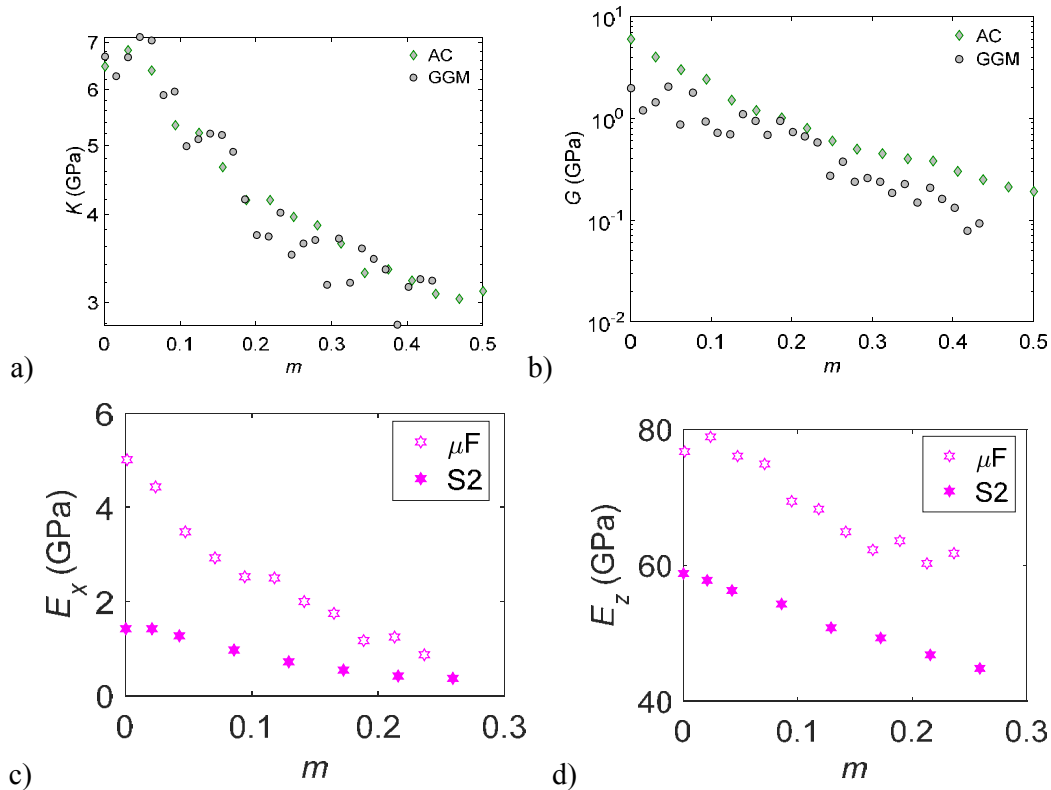


Figure 5. Mechanical properties, a) bulk ( $K$ ) and b) shear modulus ( $G$ ) of AC and GGM, and Young's modulus of  $\mu F$  and S2 ( $E$ ) c) perpendicular and d) parallel to crystalline cellulose, in function of moisture content ( $m$ ) (from Derome et al. 2018).

## CONCLUSIONS

Using Molecular Dynamics (MD) simulations, we construct and investigate atomistic models of wood polymers and of cellulose microfibril aggregates that consists of crystalline cellulose, amorphous hemicellulose and lignin. The atomistic results are upscaled and used in continuum models, using a poromechanical framework thus with full coupling of fluid transport and mechanical behavior. Upscaling to cellular scale is delineated, where the cellular scale is informed through accurate geometrical description using X-ray CT at different relative humidity.

The ensemble of results documents the full co-occurrence of sorption and swelling. This modeling methodology provides the capability to determine material properties and behavior which cannot be directly determined from experiments and to explore new pathways for material development and technology innovation, especially for capacity in terms of moisture-induced deformation.

## ACKNOWLEDGEMENT

We acknowledge the support from Swiss National Science Foundation projects no. 200021-162957 and CRSI22-125184.

## REFERENCES

Carmeliet J, Derome D, Dressler M, Guyer R. (2013). Nonlinear poro-elastic model for unsaturated porous solids, *Journal of Applied Mechanics*, 80(2), 020909.

- Derome D, Rafsanjani A, Patera A, Guyer R, Carmeliet J. (2012). Hygromorphic behaviour of cellular material: hysteretic swelling and shrinkage of wood probed by phase contrast X-ray tomography, *Philosophical Magazine*, 92: 3680-3698.
- Derome D, Rafsanjani A, Hering S, Dressler M, Patera A, Lanvermann C, Sedighi-Gilani M, Wittel FK, Niemz P, Carmeliet J. (2013). The role of water in the behavior of wood. *J. Build. Phys.* 36: 398–421.
- Derome D, Kulasinski K, Zhang C, Chen M, Carmeliet J. (2018). “Using modeling to understand the hygromechanical and hysteretic behavior of the S2 cell wall layer of wood”. In *Plant Biomechanics*, in press, edited by A. Geitmann and J. Gril. Springer.
- Hess B, Kutzner C, Van Der Spoel D, Lindahl E, 2008. GROMACS 4: algorithms for highly efficient, load-balanced, and scalable molecular simulation. *J. Chem. Theory Comput.* 4, 435–447.
- Kulasinski K, Guyer R, Derome D, Carmeliet J. (2015a). Water adsorption in wood microfibril-hemicellulose system: role of the crystalline-amorphous interface, *Journal Biomacromolecules*, 16, 2972-2978.
- Kulasinski K, Ketten S, Guyer R, Derome D, Carmeliet J. (2015b). Impact of moisture adsorption on structure and physical properties of amorphous biopolymers. *Macromolecules*, 48, 2793-2800.
- Kulasinski K, Guyer R, Derome D, Carmeliet J. (2015c). Poroelastic model for adsorption-induced deformation of biopolymers obtained from molecular simulations, *Phys. Rev. E*, 92, 022605.
- Kulasinski K, Guyer R, Derome D, Carmeliet J. (2015d) Water diffusion in amorphous hydrophilic systems: a stop and go process, *Langmuir*, 31:10843–10849.
- Kulasinski K, Derome D, Carmeliet J. (2017). Impact of hydration on the micromechanical properties of the polymer composite structure of wood investigated with atomistic simulations, *J. Mechanics and Physics of Solids*, 103: 221-235.
- The initial configuration of the polymer is constructed based on the position of atoms in the monomers and the proportion of different monomers in the chains as documented by previous studies
- Lins RD, Hünenberger PH, 2005. A new GROMOS force field for hexopyranose-based carbohydrates. *J. Comput. Chem*, 26:1400–1412.
- Oostenbrink C, Villa A, Mark AE, Van Gunsteren WF. 2004. A biomolecular force field based on the free enthalpy of hydration and solvation: The GROMOS force-field parameter sets 53A5 and 53A6. *J. Comput. Chem.* 25, 1656–1676
- Patera A, Derome D, Griffa M, Carmeliet J (2013). Hysteresis in swelling and in sorption of wood tissue. *Journal of Structural Biology*, 182:226-234.
- Patera A, Van den Bulcke J, Boone MN, Derome D, Carmeliet J. (2018). Swelling interactions of earlywood and latewood across a growth ring: global and local deformations, *Wood Sci technol.* 52:91-114.
- Rafsanjani A, Stiefel M, Jefimovs K, Mokso R, Derome D, Carmeliet J. (2014) Hygroscopic swelling and shrinkage of latewood cell wall micropillars reveal untrastructural anisotropy, *J. Royal Society Interface*, 11:20140126
- Rafsanjani A, Derome D, Carmeliet J. (2015). Poromechanical modeling of moisture induced swelling anisotropy in cellular tissues of softwoods. *RSC Advances* 5 (5): 3560-3566.